

Target Preparation for Neutron Capture Experiments on Eu Isotopes

J.M. Schwantes¹, R. Sudowe², C.M. Folden III^{1,2}, H. Nitsche^{1,2}, D.C. Hoffman^{1,2}

¹Department of Chemistry, University of California Berkeley, Berkeley, California 94720

²Lawrence Berkeley National Laboratory, Berkeley, California 94720

Neutron capture cross sections ($\sigma_{n,\gamma}$) for a variety of stable and radioactive Eu isotopes are of importance to astrophysics for understanding s-process nucleosynthesis networks and to Science-Based Stockpile Stewardship (SBSS) for interpreting radiochemical neutron fluence diagnostics. Given that theory cannot predict $\sigma_{n,\gamma}$ with sufficient precision (typically a factor of two difference between measured and calculated values is found) measurements are needed. Recently, the Detector for Advanced Neutron Capture Experiments (DANCE) located at the Lujan Center of the Los Alamos Neutron Science Center (LANSCE) has been commissioned as the state-of-art detector/facility combination to measure $\sigma_{n,\gamma}$ for neutron energies ranging from thermal (0.25 eV) to ~100 keV for a wide variety of stable and radioactive isotopes. However, suitable high-yield, high-purity target preparation methods need to be developed for these isotopes before their $\sigma_{n,\gamma}$ can be measured.

During this reporting period, the emphasis has been on preparing a radioactive target of ^{155}Eu (half-life = 4.7 years), and several stable targets, including isotopically separated ^{154}Sm (important as part of the background determination of ^{155}Eu measurements), ^{151}Eu , and ^{153}Eu . To date, a suitable backing material (beryllium) for the targets has been selected after careful calculations of its contribution to the background of the measurements. In addition, a high voltage plating procedure has been developed and optimized [1]. Stable targets of ^{151}Eu and ^{153}Eu and a target of natural Eu (~50% ^{151}Eu and ~50% ^{153}Eu) have each been plated to a mass thickness of >1 mg/cm² and delivered to the DANCE collaboration at Los Alamos National Laboratory (LANL). These targets have already been measured using DANCE and confirm that the target dimensions and backing are appropriate for performing measurements on the extremely valuable targets of separated isotopes.

In order to prepare a target of radioactive ^{155}Eu , it must first be separated from the ^{154}Sm target material that was irradiated with a neutron flux of 1.5×10^{15} neutrons/cm²/s for 50 days. The reaction is $^{154}\text{Sm} (n,\gamma) ^{155}\text{Sm}$ (half-life = 22 minutes) \rightarrow ^{155}Eu . Considerable progress has been made in developing a suitable high-yield and high-purity separation method for separating Eu from targets containing about twenty times as much Sm. A review of the literature indicated that a multiprocess approach in which Eu(III) is reduced to Eu(II) prior to separation should provide an effective and efficient means of separation from Sm(III) [1]. While all multiprocess methods identified in the literature utilize a phase change (precipitation of Eu(II)SO_4) to stabilize Eu(II) and separate it from Sm(III), co-precipitation of contaminants severely limits the purity and yield of the final product. For this reason, attempts have been made to develop a multiprocess separation method that does not require a precipitation step.

To date, a variety of multiprocess methods have been developed and tested for their ability to meet the design requirements of >99% purity and >70% yield. These methods combine an initial chemical or electrochemical reduction step using either Zn(Hg) or a potentiostat and a glassy carbon (GC) working electrode with either cation exchange chromatography and α -hydroxyisobutyrate (α -HIB) as the trivalent metal ion eluant, solvent extraction (SE) utilizing either thenoyl trifluoroacetone (TTA) or di(2-ethyl hexyl) orthophosphoric acid as trivalent ion complexants within the organic phase, or reversed phase resin chromatography (RPC) of a similar chemical nature to the organic complexants used in SE. Preliminary experiments indicate that: (a) A multiprocess approach using α -HIB as a complexing agent for trivalent lanthanides is ineffective for separating Eu from Sm because α -HIB stabilizes Eu(III) even in the presence of excess amounts of the reductant; (b) A multiprocess approach using solvent extraction shows promise, indicating that 0.1 M TTA in benzene favors extraction of trivalent over divalent metal ions by a factor of greater than 750. However, the reduction step using Zn(Hg), when combined with the TTA extraction, becomes less effective at reducing Eu during subsequent extractions and may also affect the stability of the TTA. Use of the amalgam also introduces Zn(II) contamination that must be separated from the Eu with additional solvent extraction steps. (c) An electrochemical reduction step utilizing a GC inert electrode limits contamination and is preferable over the use of the chemical reductant, Zn(Hg). However, (d) a multiprocess method combining electrochemical reduction and RPC was hindered by the presence of Cl⁻ ions that complex Eu(III). The formation of Eu(III)-chloro complexes stabilized the trivalent metal ion causing the reduction potential to become more negative, and beyond the working range of the GC electrode.

While results obtained from the current SE multiprocess method are encouraging, further refinement of a multiprocess separation method should be possible by stabilizing Eu(II) with various organic complexants such as bipyridine, phenanthroline and crown ether based ligands. Unlike inorganic complexants that are dominated by electrostatic interactions, organic ligands like these preferentially complex divalent over trivalent metal ions due to hydrophobic interactions and conformational changes that occur during chelation [2-4]. Stabilization of Eu(II) causes the reduction potential of the Eu(III)/Eu(II) couple to become more positive [4], making the reduction and the overall multiprocess separation method more effective.

REFERENCES

- [1] J.M. Schwantes, Ann. Tech. Rep., DOE/NA/00075-1 (2003).
- [2] J. Rebek, Acc. Chem. Res. 17, 258 (1984).
- [3] C.J.M. van Veggel, Chem. Rev. 94(2)279 (1994).
- [4] Y. Okaue, Nippon Kagaku Kaishi, (5)439 (1993).